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Persistent organic pollutants in the polar regions and the Tibetan Plateau: A review of current knowledge and future prospects

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ABSTRACT

Due to their low temperatures, the Arctic, Antarctic and Tibetan Plateau are known as the three polar regions of the Earth. As the most remote regions of the globe, the occurrence of persistent organic pollutants (POPs) in these polar regions arouses global concern. In this paper, we review the literatures on POPs involving these three polar regions. Overall, concentrations of POPs in the environment (air, water, soil and biota) have been extensively reported, with higher levels of dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) detected on the Tibetan Plateau. The spatial distribution of POPs in air, water and soil in the three polar regions broadly reflects their distances away from source regions. Based on long-term data, decreasing trends have been observed for most "legacy POPs". Observations of transport processes of POPs among multiple media have also been carried out, including air–water gas exchange, air–soil gas exchange, emissions from melting glaciers, bioaccumulations along food chains, and exposure risks. The impact of climate change on these processes possibly enhances the re-emission processes of POPs out of water, soil and glaciers, and reduces the bioaccumulation of POPs in food chains. Global POPs transport model have shown the Arctic receives a relatively small fraction of POPs, but that climate change will likely increase the total mass of all compounds in this polar region. Considering the impact of climate change on POPs is still unclear, long-term monitoring data and global/regional models are required, especially in the Antarctic and on the Tibetan Plateau, and the fate of POPs in all three polar regions needs to be comprehensively studied and compared to yield a better understanding of the mechanisms involved in the global cycling of POPs.

1. Introduction

The Arctic and Antarctic were for a long time assumed to be virtually devoid of anthropogenic contaminants, since these polar regions are characterized by minimal prevalence of industrial and agricultural operations. However, anthropogenic contamination in the Arctic and Antarctic ecosystems was first documented in the 1960s (Risebrou et al., 1967; Sladen et al., 1966); and since then, there has been increasing evidence showing that contaminants can reach the Arctic and Antarctic via atmospheric and oceanic transport from lower latitudes [Ahrens et al., 2010; Arctic Monitoring and Assessment Programme (AMAP) assessment report, 1998, 2004, 2009, 2015, 2016, 2017, 2018; Burkow and Kallenborn, 2000; Cabrerizo et al., 2017; Carrizo et al., 2017; Fellin et al., 1996; Gambaro et al., 2005; Gregor and Gummer, 1989; Jurado and Dachs, 2008; Kallenborn et al., 2013; Kallenborn et al., 1998; Li and Macdonald, 2005; Moeller et al., 2010; Montone et al., 2003; Nost et al., 2018; Oehme and Mano, 1984; Pozo et al., 2017; Ubl et al., 2012; Wang et al., 2017b; Wania and Dugani, 2003; Woehrnschimmel et al., 2013; Wong et al., 2018].

Among the various anthropogenic contaminants, persistent organic pollutants (POPs) are relatively more toxic, persistent, bioaccumulative and prone to long-range atmospheric transport

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(Jones and de Voogt, 1999). All these characteristics promote their widespread distribution across the globe, including regions where they have never been used (Wania and Mackay, 1993; Wania and Mackay, 1995). Additionally, when POPs travel through the atmosphere, they tend to move in stages (a repeated process of warm volatilization and cold deposition), which is known as the "hopper" effect (Jurado and Dachs, 2008). This is the reason why POPs have been found in the cold Arctic and Antarctic regions (Jurado and Dachs, 2008). Owing to the effects of POPs (toxicity and bioaccumulation) on wildlife, pesticides were first found in fat tissues of polar bears and Antarctic penguins in the late 1960s (Risebrou et al., 1967; Sladen et al., 1966). In addition to the Arctic and Antarctic, however, researchers also began to search for evidence of airborne POPs in other cold ecosystems and mountain environments (Blais et al., 1998; Chen et al., 2008; Daly et al., 2007; Grimalt et al., 2001; Morrissey et al., 2005; Wania and Westgate, 2008).

The region that encompasses the Himalaya–Hindu Kush mountain range and the Tibetan Plateau is widely known as the Third Pole (TP), with an average elevation of 4700 m. The plateau's pivotal role is due almost entirely to its height, which makes it peculiarly cold for its latitude – colder than anywhere else and containing the largest reserve of fresh water outside the polar regions (Yao et al., 2013). Thus, the similarities between the TP and the two other polar regions are their cold temperatures, remoteness, and considerable water storage. Given the close proximity to South Asia, where POPs have been extensively applied, the TP encounters more pressure from contamination by POPs (Sheng et al., 2013; Wang et al., 2008a; Wang et al., 2010a). On the other hand, the climate of the TP is more complicated and its land surface (including forest, grassland, bare soil, lakes and glaciers) is more diverse than the Arctic and Antarctic, where ice is the dominant medium. These discrepancies make the transport and accumulation of POPs in the TP region different from the two other polar regions.

Regarding research on POPs in the two main polar regions and the TP, to date (as of 2017), based on ISI Web of Knowledge, more than 2000, 300 and 100 articles can be retrieved on the Arctic, Antarctic and TP, respectively. Among these articles, 66 reviews could be found for the Arctic, two for the Antarctic, but only one for the TP region. Systematic monitoring of POPs in the Arctic has been conducted for decades; detailed information with regard to the bioaccumulation of POPs in polar bears, the risk exposure of the Arctic ecosystem to POPs, the temporal trend of POPs in the air of the Arctic, and how climate change is influencing the cycling of POPs has been successfully reviewed (Braune et al., 1999; Fromberg et al., 1999; Houde et al., 2011; Lao et al., 2017; Ma et al., 2016; McKinney et al., 2015; Riget et al., 2016; UNEP/AMAP report, 2011; Vorkamp and Riget, 2014). This body of work provides clear insights into the current and future impacts of POPs on the Arctic environment. By comparison, the long-term monitoring of POPs in the Antarctic is scarce, with most research having focused on the accumulation of POPs in biota (Baek et al., 2011; Ellis et al., 2018; Kallenborn et al., 2013; Mangani et al., 2001; Wang et al., 2017b). Then, for the TP, although a relatively limited number of studies have been conducted, there is increasing interest from scientific communities and the general public on researching POPs in this region of the world.

Given the growth in research and the associated literature in this field, we first use a science mapping method to screen and summarize the hot topics in POP research for each polar region. Intellectual milestones and the evolutionary stages of major insights and understanding are extracted. Based on the outcome of this mapping process, the aims of the study are to: (1) summarize the main outcomes and common views for the main matrix of the ecosystem, including the air, water, soil and biota; (2) address the influence of climate change on the transport and accumulation of POPs in the three polar regions, especially with respect to the release of chemicals by glacial melt; (3) synthesize the fate of POPs in the environments of the three polar regions; and (4) identify knowledge gaps and suggest where future studies should be directed.

Because AMAP - Arctic Council Working Group had published detailed assessment reports regarding fate, wildlife risk and human exposure of POPs in Arctic, this review would mainly focus on the data comparison and impact of climate change on POPs' accumulation at all three polar regions.

To the best of our knowledge, this is the first review that compiles the available achievements obtained for the two traditional polar regions, along with the TP, under the prohibition of POPs stemming from the Stockholm Convention, and provides assessments of the knowledge gaps in the scientific literature to promote best practice towards the highest quality research.

2. Summary of the hot topics in polar POP research

Pie charts (Fig. 1) were prepared based on the number of studies for different matrices over the two traditional polar regions and the TP. Research on the accumulation of POPs in biota is the dominant topic for the Arctic and Antarctic, followed air-, water-, sediment- and soil-related research. However, on the TP, POP research has focused mainly on atmospheric monitoring and transport mechanisms, with little reference to biota (Pan et al., 2014; Ren et al., 2017a; Wang et al., 2015a; Yang et al., 2011; Yang et al., 2010; Zhu et al., 2013). The temporal trends in the number of studies for the Arctic, Antarctica and TP are also shown in Fig. 1 (right-hand panel). After that the Stockholm Convention came into force in 2004 (http://www.pops.int), the international community got broadened and then commenced discussions on the need to eliminate or severely restrict POP production, thus, the amount of studies published with the aim of detecting and monitoring POPs in the Arctic and in Antarctica increased. In contrast, research on the TP started later, from 1997 (Galassi et al., 1997). This suggests that research interest has shifted from the traditional polar regions to a broader range that includes other remote areas.

Pie charts were also used to identify the important topics of wide concern among scientific researchers (Fig. 1). Regarding atmospheric POP research, the concentrations, spatiotemporal distribution and transport mechanisms (entering and moving out of the three polar regions) are the top three questions raising global concern. With respect to POPs in water, their concentrations in ice, snow, glacial runoff, remote lakes and ocean currents have been widely reported; plus, data from atmospheric monitoring and on the air–water (sea) exchange of POPs have attracted wide attention. Numerous publications have focused on the accumulation of POPs in biota, including the trophic dynamics of POPs throughout food webs, and the human exposure and health risk of POPs. Compared with research on air, water and biota, POP studies related to soils and sediments are rare, with those that have been conducted having focused mainly on the contamination of soil by POPs in the context of oil pollution in the Arctic and in Antarctica, and the related microbial diversities. However, a detailed soil survey has been conducted for the TP, in which both polluted areas in South TP and remote areas (hinterland of the Tibetan Plateau) were considered.

3. Atmosphere

Given the atmosphere is sensitive to emissions of pollutants, it reflects ongoing emissions, responds to source reductions, and acts as a carrier to disperse POPs globally (Bossi et al., 2013; Burkow and

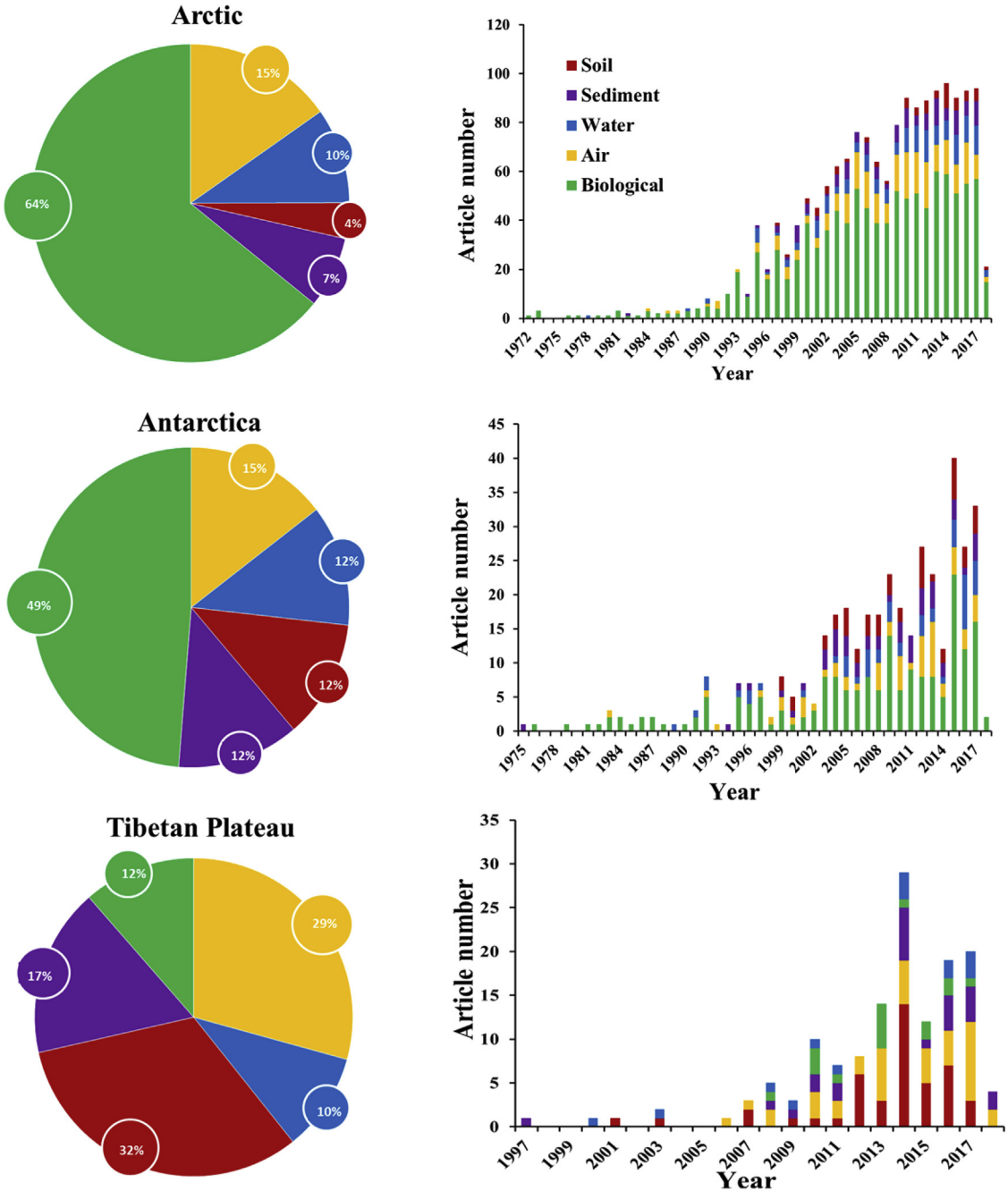

Fig. 1. Publication statistics in different years for various media in the Arctic, Antarctic and Tibetan Plateau. (note: human biomonitoing studies were included in the biota class).

Kallenborn, 2000; Gregor and Gummer, 1989; Hung et al., 2001; Kallenborn et al., 1998; Kallenborn et al., 2012; Moeller et al., 2010; Oehme and Mano, 1984; Prevedouros et al., 2004; Wania and Mackay, 1993; Welch et al., 1991; Wong et al., 2018). Thus, atmospheric monitoring is essential for the three polar regions, as it can reflect the extent of POP reduction under the regulation of the Stockholm Convention and represent how emission source regions influence remote areas (Hung et al., 2016; Kallenborn et al., 2013; Wang et al., 2017a; Wang et al., 2017b; Wang et al., 2016d). In this section, the observation stations conducting long-term atmospheric monitoring programs are summarized; and the concentrations provided by these stations are compared to identify which remote parts of the Earth are facing the greatest pressure with respect to POPs. The mechanisms and driving forces behind the atmospheric transport of POPs are then compiled.

3.1. Observation stations

There are five long-term observation stations located in the Arctic — namely, Alert [82°30′N, 62°20′ W; 200 m above sea level (masl), on Ellesmere Island, Canada], station Nord (81°36′N, 16°40′W; 24 masl, on Greenland), Stórhöfði (63°24′N, 20°17′W; 118 masl, on Iceland), Zeppelin (78°54′N, 11°53′E; 474 masl, at Ny-Ålesund on Svalbard), and Pallas (68°00′N, 24°15′E; 340 masl, in Arctic Finland) (Fig. 2a). Arctic air monitoring research was established in the early 1990s and, to date, more than two decades of

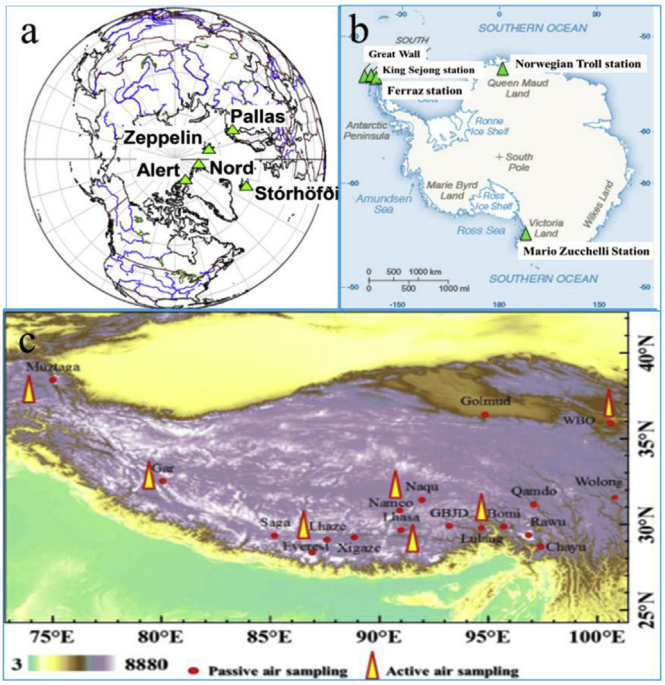

Fig. 2. Research stations for monitoring atmospheric POPs in the (a) Arctic, (b) Antarctic and (c) Tibetan Plateau.

data have been accumulated, providing invaluable information on the spatial distribution and transport processes of POPs, which can be combined with various environmental transport and fate models to assess the long-range transport of POPs.

Compared with the Arctic, the maximum consecutive sampling time for the monitoring of POPs in the Antarctic atmosphere is three years, which includes the monitoring of atmospheric polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) at the Chinese Great Wall Station on King George Island (Wang et al., 2017b, 2018), the monitoring of organochlorine pesticides (OCPs) and PCBs at King Sejong Antarctic station (Baek et al., 2011), and the monitoring of 32 PCB congeners, alpha- and gamma-hexachlorocyclohexane (HCH), trans- and cis-chlordane, trans- and cis-nonachlor, p,p'- and o,p'-DDT, dichlorodiphenyldichloroethane (DDD), dichlorodiphenyldichloroethylene (DDE), as well as hexachlorobenzene (HCB), at the Norwegian research station Troll (Dronning Maud Land) (Kallenborn et al., 2013). Besides this, atmospheric measurements of POPs have also been conducted at a monthly (Luek et al., 2017) and annual (Choi et al., 2008; Li et al., 2012a; Li et al., 2012b; Montone et al., 2003; Pozo et al., 2017) timescale, during multiple cruise-based expeditions and surveillance studies at Antarctic stations by various scientific teams. A detailed map of the sampling stations is presented in Fig. 2b.

Long-term air-monitoring observations (starting from 2008) have been conducted in the TP, but the stations are mainly located on the Tibetan Plateau, with very few observations available in North Pakistan and the Nepalese Himalaya (Sheng et al., 2013; Wang et al., 2017a; Wang et al., 2010a). Lulang is a station close to the Yarlung Tsangpo Grand Canyon (the entrance valley of the Indian monsoon), where relatively high OCP concentrations have therefore been observed (Sheng et al., 2013). Based on six years of monitoring, a recent study found a declining trend of OCPs in the atmosphere at Lulang (Wang et al., 2017a). Moreover, in 2007, a passive air-monitoring network comprising 16 sampling sites across the TP was established, which provides data on the levels of POPs at a large spatial scale, offering insights into both the spatial and temporal trends of POPs (Wang et al., 2010a; Wang et al., 2016d). Fig. 2c summarizes all the long-term and short-term air-monitoring stations located on the Tibetan Plateau.

To our knowledge, the above-mentioned 5 Arctic stations, Chinese Great Wall Station in Antarctic and Lulang station in the TP had continued POPs monitoring to date. As remote areas of the world, long-term POP observations at these stations can provide accurate data on the temporal variations of POPs, and therefore connect the linkage between the temporal patterns of POPs and climate systems.

3.2. Comparison of atmospheric POP concentrations between the Arctic, Antarctic and TP

Since the 1990s, atmospheric monitoring of POPs has been conducted at four Arctic stations – Alert, Stórhöfði, Zeppelin, and Pallas – using traditional high-volume active air samplers (AASs) (Fellin et al., 1996). Therefore, 20 years of atmospheric data on legacy POPs and PCBs, as well as 10 years of data on PBDEs, have been obtained for this region (Hung et al., 2016). Apart from this comprehensive monitoring program, other relatively short-term observations have also reported various POP concentrations in the Arctic air, including some research using passive air samplers (PASs). Detailed concentration data for legacy POPs and emerging compounds, such as perfluorinated alkyl substances, EH-TBB (2-ethylhexyl-2,3,4,5-tetrabromobenzoate), HBB (hexabromobenzene), Dec 602 (2-ethylhexyl-2,3,4,5-tetrabromobenzoate), TCEP [Tris (2-chloroethyl) phosphate], TCPP [Tris (chloroisopropyl) phosphate], TPP (Tri-phenyl phosphate), TBEP [Tris (2-butoxyethyl) phosphate], and ΣOPEs (organophosphate esters), are presented in Supporting Information (SI) Table SI-1. Similarly, the AAS data obtained from the Chinese Great Wall Station in Antarctic and other PAS data are reported in Table SI-2. The compounds include legacy POPs and PBDEs (Table SI-2).

To date, the AAS data have been gathered on the Tibetan Plateau at the stations of Lulang, Namco, Lhasa, Mt. Everest, Mt. Yulong, Nagri, and Waliguan; while PAS data cover 16 sites across the plateau (Wang et al., 2016c). Table SI-3 compiles all the reported concentrations for both legacy POPs and emerging compounds [TBE:1,2-bis(2,4,6-tribromophenoxy)ethane, DBDPE: decabromodiphenyl ethane, syn-DP: syn-dechlorane plus, anti-DP: anti-dechlorane plus, DP: dechlorane plus].

In order to compare the differences in concentrations of chemicals between the Arctic, Antarctic and the Tibetan Plateau, a bar graph showing the minimum and maximum concentrations of chemicals was prepared (Fig. 3). The results reveal the following general findings: (1) the concentrations of chemicals in the Antarctic are generally lower than those of the Arctic; (2) similar concentration ranges of HCB, α-HCH, γ-HCH and α-endosulfan are apparent between the Arctic and Tibetan Plateau; (3) wide ranges of PCB concentrations occur in the Antarctic, spanning six orders of magnitude, while the highest PCB concentrations for the three polar regions are similar, within a factor of three; (4) different from other chemicals, PBDE concentrations on the Tibetan Plateau are obviously lower than those for the Arctic and Antarctic; (5) interestingly, short-chain chlorinated paraffin (SCCP) concentrations on the Tibetan Plateau are one and two orders of magnitude higher than those of the Arctic and Antarctic, respectively (Fig. 3). Fig. 3 gives a direct insight into the levels of chemicals over the three polar regions.

3.3. Transport mechanisms

Contaminants are transported to the Arctic, Antarctic and TP by

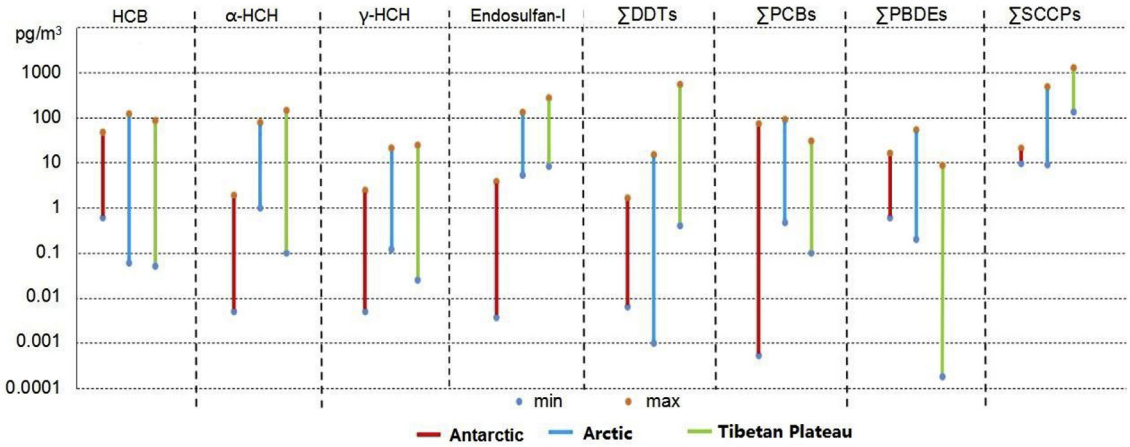

Fig. 3. Comparison of the concentration ranges of atmospheric POPs in the Antarctic, Arctic and Tibetan Plateau. Data and references used in this figure can be found in Table SI 1-3.

various pathways (Becker et al., 2011; Benskin et al., 2012b; Carrizo et al., 2017; Gong et al., 2014; Gong et al., 2015; Kallenborn et al., 2007; Kallenborn et al., 2012; Li et al., 2002; Liu et al., 2010; Moeller et al., 2010; Sheng et al., 2013; Sobek and Gustafsson, 2004), but the primary pathway is atmospheric transport. Many POPs are transported in the atmosphere to the Arctic from densely populated areas at lower latitudes, with subsequent deposition and revolatilization (Burkow and Kallenborn, 2000; Wania et al., 2017; Wania and Mackay, 1995; Wania and Mackay, 1999; Wania et al., 1999). This has been termed "the grasshopper effect" and explains how POPs travel extremely long distances and arrive in remote and cold areas (Gouin et al., 2004; Jurado and Dachs, 2008). Atmospheric transport is easily influenced by seasonal weather-related fluctuations. For example, during winter, high-pressure systems situated in Siberia push air masses north into the Arctic, which accounts for much of the Eurasian atmospheric contaminant input to the Arctic (Halsall et al., 1997). As presented by UNEP/AMAP report (2011), the climatic impact on the atmospheric transport of POPs has been investigated. ENSO (El Niño–Southern Oscillation) plays an important role in the atmospheric concentration of POPs in the Arctic. Besides, fluctuations of the AO (Arctic Oscillation) and NAO (North Atlantic Oscillation) can also change strength of the air pressures and then vary the directions of westerly winds in Arctic and Atlantic, and further change the levels and transport of POPs (Ma et al., 2004; Macleod et al., 2005; Octaviani et al., 2015). However, owing to the data on POP concentrations in the Antarctic atmosphere being relatively short term, they have yet to be linked with atmospheric circulation patterns (Kallenborn et al., 2013; Wang et al., 2017b). To date, for the TP, there are only six years of AAS POP observations available, at Lulang in the southeast of the region. Results showing almost the same seasonal fluctuation of POPs with the Indian Monsoon Index indicate that the Indian Monsoon wind drives the transport of POPs, pluming through the South Asian continent and importing POPs from there onto the Tibetan Plateau (Sheng et al., 2013; Wang et al., 2017a). Climate can influence the spatial and seasonal patterns of POPs by changing the air circulation/transport paths.

4. Vegetation

POPs can be scavenged from the atmosphere by plant leaves, the vegetation in Arctic, Antarctic and the TP is an important receptor of airborne POPs. Lichens and mosses are dominant species in Polar regions and had been considered good indicators of atmospheric pollution (Cabrerizo et al., 2012; Sanchis et al., 2015; Yogui and Sericano, 2008; Yogui et al., 2011). Banned legacy POPs, such as HCB, DDE, PCBs (Cabrerizo et al., 2012) (Yang et al., 2013) and other ongoing pollutants, such as, perfluorinalkyl substances (PFASs, Moeller et al., 2011), short-chain chlorinated paraffins (SCCPs, Lao et al., 2017), dechloranes (Yang et al., 2016) and volatile dimethylsiloxanes (VMS) (Sanchis et al., 2015) were measured in different vegetation types at three Polar regions (Augusto et al., 2013; Yang et al., 2013; Zhu et al., 2015b). Generally, lipid and wax contents of vegetation strongly affect the accumulation process, moreover, leaf morphology, physiological properties, growth rates are also responsible factors for accumulating organic air pollutants in vegetation (Franzaring and van der Eerden, 2000). Plant species with a high surface-to-volume ratio usually accumulate more organic air pollutants than species with compact leaves (Franzaring and van der Eerden, 2000). Given the portioning of organic pollutants from air to vegetation is controlled by ambient temperatures, with global warming, volatile compounds may return to the gas-phase (Barber et al., 2004). Another contrary situation is that enhanced vegetation biomass by warm temperature will provide more sorption surface for POPs' accumulation (Cabrerizo et al., 2013).

5. Ice and snow

Due to its porous structure and low temperatures, snow can effectively scavenge both gas phase and particle-bound chemicals from the air and thus mainly contribute to the atmospheric deposition of POPs in three Polar Regions. The deposition of snow POPs can be transferred deeper into aged snowpack and glacier to record the global scale long range atmospheric transport for a long time. Up to date, legacy pesticides, current use pesticides, emerging PFASs and brominated flame retardants (BFRs) had been widely detected in ice capes of Arctic [most of samplings were conducted at Svalbard, Norway (Garmash et al., 2013; Hermanson et al., 2010; Hermanson et al., 2005; Kwok et al., 2013; Ruggirello et al., 2010) and Devon Ice Cap (Meyer et al., 2012; Young et al., 2007; Zhang et al., 2013), Canada] and Antarctic[King George Island (Cipro et al., 2017) and Talos Dome (Fuoco et al., 2012)]. Because the mountain glacier at high altitude usually have limited snow melt, the deposition history of POPs can be relatively stored at high temporal resolution. In the TP, DDT and HCH records were retrieved from Mt. Everest (Wang et al., 2008a) and Dasuopu glacier (Wang et al., 2008b); PFASs temporal trends were observed from Zuoqiupu and Muztagata glacier (Wang et al., 2014a,b,c,d).

Table 1
Concentrations of POPs in ice and snow from the Arctic.

Unit		Sampling time	α-HCH	γ-HCH	∑DDTs	HCB	∑PCBs	∑PFASs	Ref.
			pg/L	pg/L	pg/L	pg/L	pg/L	pg/L	
Canadian Arctic	ice	1993					1200-6700 (3500)[a]		Gregor et al. (1995)
	ice	1993							Peters et al. (1995)
	ice	2007–2008	4.013 ± 0.307	0.423 ± 0.013					Pucko et al. (2010)
	snow	2010						260–1500	Cai et al. (2012b)
	ice	2010						2200	Cai et al. (2012b)
	snow							134–848	Veillette et al. (2012)
Central Arctic	snow	2001					2.4–6.9[b]		Gustafsson et al. (2005)
	ice	2001					2–15[c]		Gustafsson et al. (2005)
Norwegian Arctic	Ice	2006						80–200	(Kwok et al, 2013)
	snow	2003	17-382(130)	265-4390(1500)	1.73–80.5(18.8)	9.51–62.5(22.8)	160-2540 (1060)[d]		Herbert et al. (2005)
North Greenland	snow	2015	34.7–48.2						Bigot et al. (2017)

[a] $\sum_{67}$PCBs.
[b] $\sum_{15}$PCBs.
[c] $\sum_{15}$PCBs.
[d] $\sum_{14}$PCBs.

POPs concentration in snow/ice of polar regions (Table 1) and the TP (Table SI-4) were compared, displaying concentrations of HCB and DDTs in glaciers in the TP are higher than those in the Arctic; meanwhile, PCBs are lower than in the Canadian Arctic and Norwegian Arctic (Table 1 and Table SI-4). Concentration of PFASs in snow/ice cores broadly followed the order of Tibetan fresh snow > Tibetan ice core > Arctic glacier > ice cap of Antarctic (Kwok et al., 2013; Wang et al., 2014a,b,c,d; Young et al., 2007).

Temporary trends of PBDEs (Hermanson et al., 2010; Meyer et al., 2012), PCBs (Fuoco et al., 2012; Garmash et al., 2013; Zhu et al., 2015a), DDTs (Wang et al., 2008a), HCHs (Wang et al., 2008a) and PFASs (Wang et al., 2014a,b,c,d) (Young et al., 2007; Zhang et al., 2013) had been retrieved from ice cores. These trends were overall the representative of emission history of chemicals, however, due to the relocation of chemicals caused by post-depositional processes, the retrieved atmospheric deposition history exhibited some uncertainties (Pavlova et al., 2014; Zhang et al., 2013). For example, in Arctic, three PCB flux peaks were observed from 1957 to 1966, in 1974–1983, and 1998–2009 (Garmash et al., 2013). While, in Antarctic, sharp concentration increase occurred in 1930–1980, but a constant trends was from 1980 to 2002 (Fuoco et al., 2012). Both emission patterns and melting condition can lead to the above difference. An interesting phenomenon was that PCB-11 was homogeneously found in Arctic and Antarctic ice and snow (Fuoco et al., 2012; Garmash et al., 2013; Vecchiato et al., 2015), although it is not a commercial PCB product. Incineration of paper and plastics containing diarylide pigments were suggested as a major source of PCB-11 toward the Arctic (Vecchiato et al., 2015), however, its source for Antarctic is not clear.

With global warming, POPs can enter the environment of the polar regions, via the melting of glaciers and sea ice, has come to the fore (Bigot et al., 2017; Bizzotto et al., 2009; Blais et al., 2001; Guzzella et al., 2016; Meyer et al., 2009; Zhao et al., 2012; Pavlova et al., 2014). In the Arctic, high concentration episodes of β-HCH were observed throughout the year at Alert, when the Arctic sea-ice extent reached a record low. The mechanism is believed to involve re-emissions from the oceans because of sea-ice retreat sustaining levels in the Arctic air (Ma et al., 2011). Furthermore, Kwok et al. (2013) investigated the transport of PFAS from an arctic glacier to downstream locations and found melting glacier is the secondary source of PFASs. Melting glaciers can also increase the concentration of POPs in Himalayan lake water (Sharma et al., 2015). Furthermore, under global warming, POPs can migrate into lakes through the water cycle, and even accumulate in lake sediments (Schmid et al., 2011).

6. Water

Besides the air, water has become another widely used environmental matrix for monitoring POPs (Braune et al., 1999; Fernandez et al., 2005; Muir and Lohmann, 2013; Yamashita et al., 2005; Yao et al., 2002). Different from in the air, acted on by temperature, pollutants in water will migrate with phase transition (i.e., gas, liquid and solid) (Bidleman, 1999; Bigot et al., 2017; Blais et al., 2001; Cai et al., 2012a). Remote water and glaciers have been seen as "sinks" of pollutants (Gregor and Gummer, 1989; Gregor et al., 1995; Kang et al., 2009; Peters et al., 1995; Wang et al., 2014c; Wang et al., 2008b). However, in response to climate warming, oceans and glaciers possibly re-release pollutants into the environment (Jantunen et al., 2015; Ma et al., 2016; Ma et al., 2011; McKinney et al., 2015; Riget et al., 2013). In this section, the levels and spatial distribution of POPs in water in the three polar regions are compared. Furthermore, the concentrations of water pollutants reaching the polar regions are summarized, and the connections between the air–water exchange of POPs and climate warming are presented.

6.1. Concentrations

Statistically, research on POPs in ocean water for the polar regions has mainly focused on the Arctic, with twice as much work having been carried out than on the Antarctic, and ten times more than that for the TP. The concentrations of POPs in ocean water in the polar regions are summarized in Table 2 for the Arctic, and Tables SI-4 and 5 for the TP and Antarctic respectively. In general, data are still limited for the TP, not only in terms of sampling sites but also with respect to the variety of species of pollutants (Chakraborty et al., 2016; Li et al., 2017b; Ren et al., 2017b). HCHs, DDTs, and PCBs are commonly concentrated pollutants in the three polar regions (Table 1). In the Arctic Ocean, concentrations of HCHs and PCBs show wide ranges, from 0.1 to 3.2×10^3 pg/L, and from 0.13 to 4.0×10^5 pg/L, respectively, and DDTs are mainly lower than 1 pg/L (Table 2). Concentrations of HCHs and DDTs in the TP are higher than those in the Arctic and Antarctic (Table 2). Concentrations of PCBs are highest in the Arctic, followed by the Antarctic and then the TP (Table 2).

For other pollutants, the average concentration of HCB in ocean water of the Arctic is higher than that of the Antarctic (Table 2). Endosulfan and PBDEs have only been reported in water in the Arctic, with concentrations in the ranges of ND–55 and 0.3–11.2 pg/L respectively (Table 2). The concentration of the emerging

Table 2
Concentrations of POPs in ocean water from the Arctic.

Unit		Sampling time	α-HCH	γ-HCH	∑HCHs	∑DDTs	HCB	∑PCBs	∑PBDEs	Endosulfan	∑PFASs	OPEs	Ref.
			pg/L	pg/L	pg/L	pg/L	pg/L	pg/L	pg/L	pg/L	pg/L		
Arctic Ocean	surface water	1993		250						3.8			(1)
	deep water	2001, 2005, 2008						0.13–21[a]					(2)
	deep water	2001, 2005, 2008							0.3 –11.2[g]				(3)
	deep water	2001, 2005, 2008				0.10–66							(4)
	surface water	2004						<1[b]					(5)
	surface water	2008–2009			262–3156	ND-0.31				0.04–0.18			(6)
		2008	630-2690 (1978)	170-700 (382)						ND-55			(7)
		2008	0.12–0.76 (0.44)	0.20–0.37 (0.26)	0.1–5.14 (1.15)	ND-0.16 (0.09)				ND-0.1 (0.05)			(8)
	surface water	2010											(9)
Canadian Arctic	2 m	1998	1100 ± 7 00	200 ± 10	1400 ± 100								(10)
		2005–2009											(11)
	7 m	2007–2008	465–1013	150–254			4.0–6.4						(12)
		2007, 2008, 2010								0.20–2.3			(13)
											27–754		(14)
												DL-4400	(15)
		2010–2011									1900 –153000		(16)
Greenland	11 m	2009									14.5–449		(17)
	11 m	2009									50–280		(18)
	5 m	2010	11.00				50.00						(19)
		2015	224–253										(20)
Europe Arctic	8 m	2004	ND-65 (13)	ND-21 (4.7)	1.5–83 (16.9)	0.17–0.96 (0.4)	0.8–9.6 (4.6)						(21)
	8 m	2009							0.03 –0.64 [h]				(22)
Norwegian Arctic	surface water	2009						2000 –400000[c]					(23)
		2009						11.6–27.7[d]					(24)
Russian Arctic	5 m	2003–2005			291	3	9	12 [e]	5 [i]				(25)
Western Arctic	surface water	2010									500 ± 170		(26)
Eastern Arctic		1996	910	270									(27)
Central Arctic	8 m	2001						0.13–3.92 (1.34)[f]					(28)
	surface water	2012									10–1700		(29)
Bering Sea and Chukchi Sea		1999			226–652								(30)

(1)(Weber et al., 2006); (2)(Carrizo and Gustafsson, 2011a,b); (3)(Salvado et al., 2016); (4)(Carrizo et al., 2017); (5)(Gioia et al., 2008); (6)(Qiu and Cai, 2010); (7)(Jantunen and Bidleman, 1998); (8)(Cai et al., 2010); (9)(Ke et al., 2017); (10)(Moisey et al., 2001); (11)(Benskin et al., 2012b); (12)(Wong et al., 2011); (13)(Morris et al., 2016); (14)(Veillette et al., 2012); (15) (Mcdonough et al., 2018); (16)(Lescord et al., 2015); (17)(Busch et al., 2010); (18)(Zhao et al., 2012); (19)(Carlsson et al., 2012); (20)(Bigot et al., 2017); (21)(Lohmann et al., 2009); (22)(Moeller et al., 2011); (23)(Polkowska et al., 2011); (24)(Papale et al., 2017); (25)(Carroll et al., 2008); (26)(Cai et al., 2012b); (27)(Harner et al., 1999); (28)(Sobek and Gustafsson, 2004); (29)(Yeung et al., 2017); (30)(Yao et al., 2003).

[a] $\sum_{13}$PCBs.
[b] $\sum_{6}$PCBs.
[c] $\sum_{7}$PCBs.
[d] $\sum_{9}$PCBs.
[e] $\sum_{10}$PCBs.
[f] $\sum_{12}$PCBs.
[g] $\sum_{14}$PBDEs.
[h] $\sum_{10}$PBDEs.
[i] $\sum_{4}$PBDEs.

group of compounds, PFASs, in Arctic water, is comparable to that of King George Island but higher than that of the western Antarctic (Table 2).

6.2. Spatial distribution

Owing to studies on POPs in water in the Antarctic and TP being quite scattered, examining the spatial distribution of POPs in water has instead focused mainly on the Arctic. The Arctic Ocean is composed of the central Arctic Ocean and seven vast continental shelf seas (including the Chukchi, East Siberian, Laptev, Kara, Barents, Beaufort and Lincoln seas) that cover 53% of its waters (Figure SI-1) (Jakobsson, 2002). In the Arctic, large-scale observations have also been performed along the cruise tracks of Arctic Ocean expeditions (Benskin et al., 2012a; Cai et al., 2010; Cai et al., 2012b; Jantunen and Bidleman, 1998; Lohmann et al., 2009; Moeller et al., 2010; Sobek and Gustafsson, 2004; Xie et al., 2011; Yao et al., 2002). Overall, the concentrations of POPs in surface water have shown decreasing trends with increasing latitude, which is consistent with distances away from source regions (Benskin et al., 2012b; Carrizo et al., 2017; Gioia et al., 2008; Salvado et al., 2016; Sobek and Gustafsson, 2004; Yao et al., 2002). Longitudinally, the values of POPs tend to decrease from west to east, but this trend is not statistically significant (Yao et al., 2002). For different parts of the Arctic Ocean, the concentrations of POPs in the water of shelf-sea areas are usually higher than in the central Arctic ocean (Carrizo and Gustafsson, 2011a,b; Carrizo et al., 2017; Salvado et al., 2016; Sobek and Gustafsson, 2004).

Dissolved contaminants may reach deeper water layers through vertical mixing or convective overturning in well-mixed shelf areas (Carrizo et al., 2017; Sobek and Gustafsson, 2014a). The vertical profile of dissolved-phase POPs in surface seawater shows a general decreasing trend in concentration with depth, such as for DDT, HCB, Endosulfan, and the low-molecular-weight PCBs, and PBDEs, in the Arctic Ocean (Cai et al., 2010; Fernandez et al., 2005; Salvado et al., 2016; Sobek and Gustafsson, 2014a; Sun et al., 2016; Yamashita et al., 2005; Yamashita et al., 2008). However, in deep water, increasing concentrations of DDE and high-molecular-weight compounds with depth have been observed (Carrizo et al., 2017; Salvado et al., 2016; Sobek and Gustafsson, 2014b). For the vertical distribution of emerging compounds, the detection of PFAS has been shown to be limited to the polar mixed layer (15–30 m) and halocline (150 m below the surface); whereas, in the North Barents Sea, perfluorooctane sulfonate (PFOS) has been detected down to 250 m below surface (Yeung et al., 2017).

6.3. Oceanic and riverine transport

Aside from atmospheric transport, both water-soluble and particle-bound compounds can be transported via rivers or ocean currents (Alexeeva et al., 2001; Carroll et al., 2008; Sun et al., 2016). For example, HCH is generally regarded as a “swimmer”, being stored in rivers and ocean water; plus, owing to the “wash-out” of rivers, large areas of land can be flushed into the Arctic, providing an extra release of POPs into this region (Alexeeva et al., 2001; Carlsson et al., 2012; Carrizo and Gustafsson, 2011a,b; Carroll et al., 2008; Lohmann and Belkin, 2014; Nikanorov et al., 2015; Zhulidov et al., 2000; Zhulidov et al., 2002). Along with transport via oceanic currents, phytoplankton can also carry POPs into the oceans (Dachs et al., 2002; Galban-Malagon et al., 2012) – a mechanism referred to as the “biological pump”. In the Greenland current and Atlantic sector, the transport of PCBs and HCHs to the Arctic Ocean is strongly retarded by the oceanic biological pump (Galban-Malagon et al., 2012; Galban-Malagon et al., 2013). High settling fluxes of PCBs due to the biological pump have also been observed in the Southern Ocean (Galban-Malagon et al., 2018).

6.4. Air–water exchange and its relationship with climate change

Gaseous exchange occurs constantly for POPs at the air–water interface (Bidleman, 1999; Jantunen and Bidleman, 1996; Jantunen et al., 2008; Wong et al., 2011). However, air–water gas exchange is a two-way process, which can alternate between net deposition and net volatilization in response to seasonally changing temperatures, atmospheric levels, cycles of biological productivity, and so on. Currently, the seasonal variations of air–water exchange are observed in three polar regions, but long-term trends are only available for the Arctic. In summer, HCB, γ-HCH and β-HCH are close to equilibrium or undergoing net deposition, with occasional reports of volatilization in some areas or seasonal variations in the polar oceans and lake water of the TP (Bidleman et al., 2007; Bigot et al., 2016; Cai et al., 2012a; Cincinelli et al., 2009; Ding et al., 2007; Ren et al., 2017b). Currently used pesticides, such as α-endosulfan, are generally dominated by net deposition (Weber et al., 2006).

Based on long-term data in the Arctic, the connection between air–water exchange and climate change has been explored using both observational data and models (Jantunen et al., 2015; Ma et al., 2016; Ma et al., 2011; McKinney et al., 2015; Xie et al., 2011). The direction of α-HCH shifted from net deposition in the 1980s to net volatilization in the 1990s (Fig. 4), which was influenced by declining primary emissions and the climate warming (Cai et al., 2012a; Ding et al., 2007; Jantunen and Bidleman, 1996; Jantunen and Bidleman, 1995; Jantunen et al., 2008). Since 1990, the net direction of α-HCH gaseous exchange has likely been controlled more by global warming and rapid changes in sea-ice coverage than by changes in global emissions of α-HCH (Cai et al., 2012a; Wong et al., 2011). Based on an atmospheric model for POPs, Ma et al. (2011) they found that other toxic chemicals (such as p,p'-DDT, PCB congeners 28, 52 and 101) probably volatilize from secondary emission sources/reservoirs in water, snow and ice across the Arctic under climate warming (Ma et al., 2011). Therefore, under Arctic warming, some POPs in water will return back to the atmosphere, becoming available for circulation once again, which will in turn offset the effect of reducing the primary emission sources.

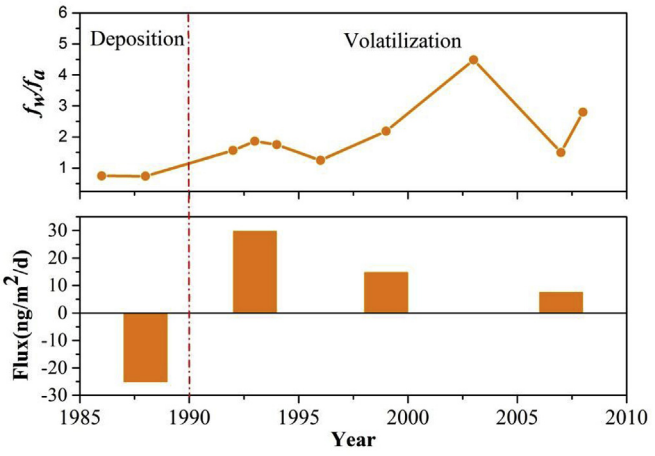

Fig. 4. Temporal trends for air–water gas exchange fugacity ratios and flux of α-HCH in the Arctic Ocean [data are from Cai et al. (2012a), Ding et al. (2007), Jantunen and Bidleman (1996), Jantunen and Bidleman (1995) and Jantunen et al. (2008)].

7. Soil and sediment

Soil and sediment in the cold polar regions and high mountains provide stationary "end points" where POPs possibly reside for the long term (Borghini et al., 2005b; Chen et al., 2008; Wania and Mackay, 1993; Wania and Westgate, 2008). Without obvious local emission sources, soil and sediment in the polar regions act as natural recorders of POP levels over long timescales, thus accumulating valuable information on past environmental and climatic events.

7.1. Soil

Comparisons of POPs in the soils of the three polar regions are summarized in Table 3, from which it is clear that the levels/orders of magnitude are similar to those mentioned above for water. The spatial distribution of POPs in soil in the Arctic and Antarctic has mainly been examined on small scales (specific islands) (Borghini et al., 2005a; Cabrerizo et al., 2013; Casal et al., 2018; Klanova et al., 2008; Negoita et al., 2003; Park et al., 2010; Wang et al., 2012a; Wang et al., 2015b). Whereas, studies on POPs in soil in the TP have been carried out across the plateau on a larger scale (Tao et al., 2011; Wang et al., 2014a; Wang et al., 2012c; Zheng et al., 2012). Generally, concentrations show decreasing trends from the edge to the hinterland areas of the TP, and the profile is dominated by more volatile compounds (Klanova et al., 2008; Park et al., 2010; Wang et al., 2012a; Wang et al., 2012b; Wang et al., 2014a; Wang et al., 2012c), which reflects well the long-range atmospheric transport of POPs. Besides, higher levels of POPs have been observed sporadically near research stations (Abakumov et al., 2014; Aislabie et al., 1999; Li et al., 2017a; Park et al., 2010; Wang et al., 2014a; Wang et al., 2016b) and military bases (Bright et al., 1995; Stow et al., 2005) in the polar regions or populated regions of the TP, indicating local sources at these sites.

Soil in cold regions are considered as "sinks" of POPs (Meijer et al., 2003), but the direction of air–soil gas exchange of POPs may change with variations in temperature and soil organic matter (SOM) (Wang et al., 2012c). Cabrerizo et al. (2013) provided strong evidence that both climate change (temperature) and carbon cycling (SOM content) drive the remobilization of PCBs in the Antarctic, not only due to warming. In the TP, grassland and forest soil are sinks of DDTs, while desert soil might re-release low-molecular-weight PCBs, induced by variations in SOM (Wang et al., 2014b; Wang et al., 2014d; Wang et al., 2012c).

7.2. Sediment

Sediments are recognized as a useful record of atmospheric deposition (Fernandez et al., 2000; Malmquist et al., 2003; Martins et al., 2010; Rawn et al., 2001; Sun et al., 2005; Zaborska et al., 2011). The geographical distributions of POPs in surface sediment are usually similar to those for soil or water; plus, the temporal variations of POPs vertically in sediments provide a record of their emissions history globally (Cheng et al., 2014a; Cheng et al., 2014b; Combi et al., 2017; Dong et al., 2015; Duan et al., 2011; Galassi et al., 1997; Guo et al., 2011; Jin et al., 2017; Jin et al., 2015; Li et al., 2017b; Lin et al., 2017; Ma et al., 2017a; Ma et al., 2017b; Malmquist et al., 2003; Sobek and Gustafsson, 2014a; Sun et al., 2018; van Drooge et al., 2013; Wang et al., 2010b; Yang et al., 2016). Both of these aspects have been extensively reported and will not be reviewed here. Instead, in this section, the shifts in the sources of POPs in sediments over long timescales, along with the relationships between POP levels and sedimentary characteristics, are the focus.

Studying the temporal variation in the concentrations and relative compositions of POPs has generally been the main approach to tracing the possible sources of such pollutants (Blais et al., 2001; Dahle et al., 2003). Against the background of climate warming, glacial melt has become another dominant source of POPs for nearshore waters adjacent to glaciers and lakes (Bigot et al., 2017; Blais et al., 2001; Cai et al., 2012b; Sharma et al., 2015). In the sediments of glacial lakes, a second peak of DDTs and HCHs was observed in the 1990s after the ban of these compounds (Cheng et al., 2014a; Li et al., 2017b; Sun et al., 2018) – a phenomenon believed to derive from the melting of glaciers (Blais et al., 2001).

Besides sources, sedimentary properties [including grain size, water content, loss on ignition, total organic content (TOC), and black carbon] might play a role in the vertical distribution of POPs (Fernandez et al., 1999; Hong et al., 2012; Li et al., 2017d; Ma et al., 2015; Xue et al., 2016). Thus, the relationships between POPs and sedimentary properties have been explored in several studies. For example, no apparent co-relationships were obtained between PCB concentrations and the abovementioned sedimentary properties for the Bering Sea, Chukchi Sea, and Canada Basin (Hong et al.,

Table 3
Comparison of POPs in soils from the polar regions and the Tibetan Plateau (TP).

Sampling sites	Sampling time	∑HCHs pg/g	∑DDTs pg/g	HCB pg/g	∑PCBs pg/g	∑PBDEs pg/g	Ref.
Arctic							
Ny-Ålesund	2013					42	Wang et al. (2015b)
Ny-Ålesund	2007						Wang et al. (2009b)
Island of Tromsø	2013–2014	NA-81		616–1477	300–950		Casal et al., (2018)
Antarctic							
Livingston Island	2009				24.5–40.9		Cabrerizo et al., (2013)
Eastern coast of Antarctica	1998	86–469	110–1220	20–25280	200–410		Negoita et al., (2003)
King George Island	2006				8.0–33.8		Park et al., (2010)
Tibetan Plateau							
Tibet		670–5380	ND-2830				Fu et al., (2001)
Mt. Qomolangma	2005				47.1–422.6	4.3–34.9	Wang et al. (2009a)
Tibetan plateau	2005–2006	467 ± 741	329 ± 818				Tao et al., (2011)
	2007	64–847	13–7700	24–564	75–1021	ND-27	Wang et al. (2012c)
	2008				163	26	Zheng et al., (2012)
	2011						Wang et al., (2013)
	2007						Wang et al. (2014a)
	2014	430–5690	21–1230				Tao et al., (2014)
Maizhokunggar County	2015						Li et al. (2017c)
Western TP	2014						Zhou et al., (2018)

2012), and grain size was found to be a minor factor for the accumulation of POPs (Li et al., 2017d; Xue et al., 2016). However, significant relationships of PBDEs with total organic carbon (TOC) and black carbon have been found (Cai et al., 2012c; Xue et al., 2016). These observations indicate that, besides sources, the distribution of POPs in sediment is controlled by multiple factors, such as atmospheric transport and deposition, mixing, partitioning, and sorption in the water column and sediments.

8. Biota

Due to lipophilicity, POPs can bioaccumulate in organisms and are stored in the lipid component of tissues (Muir et al., 1988). The bioaccumulation and biomagnification of POPs along aquatic and terrestrial food chains have been widely reported (Corsolini et al., 2017; Corsolini et al., 2003; Corsolini et al., 2002; Corsolini and Sara, 2017; Evenset et al., 2016; Goerke et al., 2004; Hop et al., 2002; Kelly and Gobas, 2001; Kelly and Gobas, 2003; Lescord et al., 2015; Morris et al., 2016). Because of their distance from emission sources, biota in the polar regions can serve as adequate biological indicators of global POP emissions (Braune et al., 1999; Braune, 2007; Braune et al., 2002; Braune et al., 2015; Braune and Mallory, 2017; Helgason et al., 2008; Riget et al., 2010a; Riget et al., 2013; Riget et al., 2010b). To date, the levels, bioconcentration mechanisms, and temporal trends of POPs in biota from the polar regions have been extensively reported and reviewed by other papers (Braune et al., 1999; Cannon et al., 2016; Corsolini, 2009; Houde et al., 2011; Letcher et al., 2010; McKinney et al., 2015; Vorkamp, 2016) and AMAP assessment reports (2009, 2015, 2018). Here, the factors influencing the levels and bioaccumulation of POPs in biota in the polar regions, as well as their long-term trends and exposure effects, are reviewed.

8.1. Factors influencing levels and bioaccumulations

The levels of POPs in the Arctic, Antarctic and TP vary by region, biota type, and even tissue type (Braune et al., 1999; Braune et al., 2002; Braune et al., 2005; Corsolini et al., 2003; Court et al., 1997; Inomata et al., 1996). For example, differences have been observed between benthic and pelagic species (Goutte et al., 2013; van den Brink et al., 2011). Goutte et al. (2013) found levels of HCB to be far higher in pelagic than in benthic organisms of Antarctic. Also, concentrations in marine biota have been found to be higher than those in freshwater and terrestrial ecosystems (Dietz et al., 2000). Among the different tissues, liver tissue, with its relatively higher lipid content, accumulates higher concentrations of POPs (Aas et al., 2014; Andersson et al., 1998).

Biological (lipid dynamics, food-web productivity and migration) and environmental (emissions and climate change) factors possibly play a role in bioaccumulations of POPs in the polar regions (Andersson et al., 1998; De Laender et al., 2010; Fisk et al., 2001; Gustayson et al., 2015; Li et al., 2016; Pedro et al., 2017; Rios et al., 2017). In response to the limited observational data available in the polar regions, models have been set up to examine to what extent these factors cause bioaccumulation in biota (Borga et al., 2010; De Laender et al., 2010; Kelly and Gobas, 2003). For example, driven by lipid dynamics, “bioaccumulation factors” in fish were found to be 10 times lower in summer than in spring and fall/winter (De Laender et al., 2010); and driven by seasonal shifts in trophic levels and lipid dynamics, “trophic magnification factors” were highest for PCB 153 during spring and lowest for PCB 52 in summer and fall/winter (De Laender et al., 2010). Furthermore, increased temperatures and emissions were found to reduce the overall bioaccumulation of organic contaminants in the Arctic marine food web, with the largest change being for PCB-52 and PCB-153 (Borga et al., 2010). For the less water-soluble compounds, increased emissions reduced the bioavailability, and increases in temperature resulted in an overall reduction in net bioaccumulation (Borga et al., 2010).

8.2. Long-term trends

Biota data in the polar regions were collected from literature that covered different time periods and different statistical analysis methods to build long-term trends (Cincinelli et al., 2016; Ellis et al., 2018; Helgason et al., 2008; Riget et al., 2010a; Riget et al., 2019). The longest time span involved the reporting of POPs in penguins for nearly 50 years (1964–2011) in the Antarctic (Ellis et al., 2018), but other long-term studies (over 20–30 years) have also been conducted (Cincinelli et al., 2016; Goerke et al., 2004; Helgason et al., 2008; Riget et al., 2010a). The biota chosen to reflect the long-term trends of POPs mainly include eggs, fat tissues in penguins, fish, and *Trematomus bernacchii* (AMAP, 2016; Braune, 2007; Ellis et al., 2018; Goerke et al., 2004; Helgason et al., 2008; Riget et al., 2010a; Riget et al., 2010b, 2019). Generally, legacy POP levels in most species increased from the 1960s–1980s, and then decreased from the 1990s to the present day, with only very few cases of significantly increasing trends having been reported (Braune and Letcher, 2013; Cincinelli et al., 2016; Ellis et al., 2018; Goerke et al., 2004; Helgason et al., 2008; Riget et al., 2010a).

The temporal trends of POPs in biota are influenced by the expression of their concentrations, of food webs (pelagic or benthic), and of species and locations (Evenset et al., 2016; Goutte et al., 2013; Lescord et al., 2015; Moisey et al., 2001; Riget et al., 2010b). For example, Riget et al. (2010b) found decreasing trends of POPs during the period 1994–2008 in Arctic Char became non-significant when expressed by lipid weight rather than wet weight. Significantly increasing concentrations within a decade (1987–1996) were observed for two benthic fish species, while the pelagic species did not increase (Goerke et al., 2004). van den Brink et al. (2011) concluded that concentrations of PCBs, DDE, HCB and dieldrin were declining during 1993–2003 in Antarctic pelagic seabirds, but not in benthic organisms. Braune (2007) measured OCPs and PCBs in eggs of three species (murres, fulmars and kittiwakes) in the Arctic, with the temporal trends for HCH (1975–2003) found to be different among the species. Riget et al. (2010a) observed that the highest mean decreasing rate in Arctic biota was for α-HCH, γ-HCH and DDTs, while the lowest rate was for total chlordanes and toxaphene. Moreover, β-HCH showed a low or nonexistent rate of decline in the western Arctic, but greater decreasing trends to the east (Riget et al., 2010a).

8.3. Exposure assessment

The bioconcentration and toxicity of POPs result in high exposure risks for species at the top trophic levels (Allen-Gil et al., 1997; Andersen et al., 2001; Braune et al., 2005; Bustnes et al., 2007; Goerke et al., 2004; Hoekstra et al., 2003; Hop et al., 2002; Kelly and Gobas, 2001; Kelly et al., 2008; Rudolph et al., 2016; Villa et al., 2017). Moreover, the natural stresses on biota in the extreme environments of the polar regions are often more severe than in other regions (https://www.amap.no/documents/doc/amap-assessment-2018-biological-effects-of-contaminants-on-arctic-wildlife-and-fish/1663, AMAP, 2018), rendering species here more vulnerable to the effects of pollutants.

Comparatively, the Arctic has attracted more attention, due to the traditional lifestyle of the Inuit population appearing to be an important contributor to POP exposure (Dallaire et al., 2009; Valera et al., 2013; Zhulidov et al., 2002). Letcher et al. (2010) and https://www.amap.no/documents/doc/amap-assessment-2015-human-

health-in-the-arctic/1346, AMAP (2015) reviewed the biological effects in relation to organohalogen contaminant exposure in several top trophic level species of the Arctic, and tried to assess known tissue/body compartment concentration data in the context of possible threshold levels of effects to evaluate the risks. Dallaire et al. (2009) found the consumption of fish and marine mammals appears to be an important contributor to PFOS and PBDE exposure among Nunavik Inuit. Valera et al. (2013) concluded that some PCB congeners and p,p'-DDE are associated with a higher risk of hypertension in Inuit adults.

Only small amounts of exposure-assessment research in the Antarctic and TP have been reported (Borghesi et al., 2008; Wang et al., 2016b). In the Antarctic, the TEQ values of PCBs and OCPs, and the "toxic equivalent factors" of PCDF congeners, have been used to estimate exposure risks (Borghesi et al., 2008; Schiavone et al., 2009). The TEQ values were found to be one order of magnitude lower than those considered to elicit physiological effects in aquatic mammals (Borghesi et al., 2008; Schiavone et al., 2009). Generally, POP concentrations are not currently a risk in terms of generating adverse health effects. In the TP, the exposure risks with respect to POPs have mainly been examined in the context of human beings Bi et al. (2016); Chen et al. (2015); Gong et al. (2011); Liu et al. (2014); Pan et al. (2014); Pokhrel et al. (2018); Wang et al. (2016a); Wang et al. (2016b). The results showed that OCPs in farmland areas do not pose any non-carcinogenic risks, and only pose a very low cancer risk to Tibetan residents (Wang et al., 2016b).

9. Modeling

The presence of POPs in the polar regions derive primarily from external inputs, as evidenced by the numerous studies mentioned above, for all environmental media (atmosphere, water, soil, sediment, and biota). However, although the transport pathways of POPs from source regions to the polar regions have been observed, the specific mechanisms of transportation and the environmental burdens of these substances are harder to determine based on observational data alone. Thus, models involving multi-compartmental cycling are needed to simulate the transport mechanisms and verify the fate of POPs in the polar regions. Several models have been developed to examine the transport and transfer of POPs from low latitudes to the Arctic, but very few have been developed to investigate the equivalent the Antarctic and TP.

Based on the direction of transport (i.e., horizontal and vertical), both 2-D and 3-D transport models have been developed (Christensen, 1993; Krapivin, 1995; Wania and Mackay, 1995). Models concerning the transport of POPs to the Arctic are as follows: (1) global atmospheric distribution models for POPs (Wania and Dugani, 2003; Wania and Mackay, 1993; Wania and Mackay, 1995; Wania and Mackay, 1999; Wania et al., 1999); (2) the Danish Eulerian Hemispheric Model (Friedman and Selin, 2016; Hansen et al., 2004); (3) the Arctic Mass Balance Box Model (Li et al., 2004); GEM/POP (Huang et al., 2007); (4) ocean transport model (Armitage et al., 2006; Armitage et al., 2009; Stemmler and Lammel, 2009; Stemmler and Lammel, 2010; Stemmler and Lammel, 2013; Zhang et al., 2017); (5) a coupled atmosphere–ocean general circulation model (Guglielmo et al., 2009); (6) a 2-D atmospheric transport model (Ma, 2010); (7) a 3-D atmospheric transport model (Zhang et al., 2010); and (8) GEOS-Chem (Friedman and Selin, 2012; Friedman and Selin, 2016).

Some atmospheric transport models have been used to estimate the transmission height and distance for POPs from source regions (Asia, Europe and North Atlantic) to the Arctic (Halsall et al., 2001; Ma, 2010; Zhang et al., 2010; Octaviani et al., 2015). For example, the poleward atmospheric transports of γ-HCH from Asian or Eurasian sources to the Arctic have been found to occur mostly in the mid-troposphere (e.g., from 3000 m to 5500 m) (Zhang et al., 2010). Huang et al. (2007) used GEM/POPs to demonstrate that, for PCBs, transport flux peaking occurs below 8 km for the gaseous phase and 14 (6) km for particulate PCB-28 (PCB-180).

The transport mass of POPs in the Arctic has also been estimated by models. For instance, Huang et al. (2007) estimated the Arctic receives 5914 kg (89 kg) of PCB-28 (PCB-180) from Europe, 1217 kg (26 kg) from the North Atlantic, and exports 2413 kg (8.7 kg) of PCB-28 (PCB-180) to North America, and 21.4 kg PCB-180 to Asia in year 2000. Between 1945 and 2000, the total loading of α-HCH to the Arctic Ocean was approximately 27 700 t, accounting for approximately 0.6% of all global α-HCH emissions from agricultural land to the atmosphere (Li et al., 2004). Similar results were reported by Wania and Mackay (1999), who found only very small fractions of α-HCH (<1%) have reached the Arctic. The dominant input pathways for α-HCH entering the Arctic are gas absorption (50%) and ocean currents (34%); and the major means of removal of α-HCH are microbial degradation (49%) and ocean currents (23%) (Li et al., 2004).

Climate change also impacts the atmospheric transport and fate of POPs in the Arctic (Hansen et al., 2015; Lamon et al., 2009; Ma et al., 2016; Ma et al., 2011). By the end of the 2090s, the mass of HCHs within the Arctic is predicted to be up to 38% higher than that in the 1990s, whereas the change in the mass of PCBs is expected to range from 38% lower to 17% higher (Hansen et al., 2015). Emerging contaminants, i.e., those with no or only a short emissions history, will be more rapidly transported to, and build up within, the Arctic environment in the future warmer climate (Hansen et al., 2015). Considering the influence of climate- and emissions-driven processes on Arctic PCB concentrations, Friedman and Selin (2016) evidenced that the emission or removal/transformation processes occurring outside the Arctic, rather than within, have a greater influence on atmospheric PCB levels.

10. Research needs and future perspectives

Although the levels of atmospheric POPs have been reported for the polar regions, the temporal trends and half-lives for specific compounds, and the role that atmospheric circulation plays in the transport of "emerging" POPs, remain unclear for the Antarctic and the TP. Thus, long-term and consistent monitoring of atmospheric POPs is required in the polar regions.

The monitoring of POPs in water has yielded data for more than 20 years in the Arctic and Antarctic, whereas related work in the TP has only just begun. The TP is also often referred to as the "Asia water tower", because it is home to the most mountain glaciers in the world, as well as thousands of lakes on the Tibetan Plateau, both of which are vital to Asia water security and beyond. In this context, more observations of POPs in water in the TP are needed to evaluate the global cycling of POPs.

Systematic research has been performed on the levels and bioaccumulation of POPs in the aquatic food webs of the polar regions. Comparatively, however, fewer data have been published on terrestrial food chains, both in terms of the quantity and length of the chains, because only two short terrestrial food chains (lichen—caribou—wolf in the Arctic and air—grass—yak in the TP) have thus far been reported. Due to close association that terrestrial food chains have with human life, more work on the bioaccumulation mechanism from the environment to higher trophic levels is urgently needed in the future.

So far, results have indicated that POPs in the Antarctic and TP do not cause adverse effects on animals or humans; whereas, in the Arctic, the traditional lifestyle of the Inuit population has not protected them against exposure to POPs, particularly emerging

pollutants. Continuous attention should be paid to the health risk of POPs on the Inuit and Tibetan people. Regardless, there remains a dearth of data on the true nature of contaminant exposure, especially the cause–effect relationships with respect to the exposure of these contaminants in wildlife and people.

Models have been used to simulate the global transport of POPs to the Arctic, but models that simulate the transport processes and masses of POPs in the Antarctic and TP should be developed. Moreover, it will be challenging to develop models that cover the complexity of the transport processes (including glacial melt) for POPs across the Himalaya and in regions that experience strong solar radiation.

Climate change and emissions reduction measures act jointly on the fate of POPs. In this regard, the polar regions are highly sensitive to climatic factors but far away from direct emission sources, thus offering a natural laboratory to carry out research on the effects of both climate and emissions on the fate of POPs. However, presently, studies are limited by a lack of long-term and consistent data, making it difficult to fully understand the mechanisms involved, especially in the Antarctic and TP. Hence, the long-term monitoring of POPs together with the development of emissions inventories are a necessary next step.

Finally, although the environmental processes related to POPs have been separately observed in each polar region, there remains no consensus on the fate of POPs in remote and cold regions. Associated work in the three polar regions on all environmental media (air, soil, water and biota) should be placed on the agenda.

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Appendix A. Supplementary data

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